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Ozone/peroxide advanced oxidation in combination with biofiltration for taste and odour control and organics removal

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HIGHLIGHTS

- BAC alone may serve as a suitable treatment technology for T&O control at warmer temperatures.
- Biologically activated anthracite outperformed non-biological anthracite in removing both geosmin & MIB.
- O_3 efficiency and BAC biodegradation rates significantly decreased in cold water conditions.
- H_2O_2/O_3 advanced oxidation improved MIB removal in cold water conditions.
- Biopolymers were observed to preferentially react with $OH rather than O_3.

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ABSTRACT

The objective of this pilot-scale study was to investigate the effectiveness of incorporating ozone (O_3) and advanced oxidation (hydrogen peroxide/ozone: H_2O_2/O_3) in combination with biofiltration for taste and odour control, organic carbon removal, and disinfection byproduct (DBP) precursor reduction. Implementation of O_3 and H_2O_2/O_3 with and without biofiltration was investigated at pilot-scale in terms of geosmin, 2-methylisoborneol (MIB), and DBP precursor removal efficiency. Two media types (granular activated carbon and anthracite) were compared in conjunction with investigating the impact of pre-oxidation with O_3 (2 mg/L) and varying H_2O_2/O_3 mass ratios (0.1, 0.2, 0.35, and 0.5 mg/mg). When O_3 preceded biologically active carbon (BAC) or biologically active anthracite, geosmin removals of 80% and 81%, respectively, were observed at 10 °C; this increased to 89% and 90%, respectively, at 16 °C. Optimal MIB removal (67%) was achieved with 0.1 H_2O_2/O_3 (mg/mg) in combination with BAC at 16 °C.

In general, geosmin proved to be more amenable to biodegradation than MIB. BAC without pre-oxidation removed 87% geosmin and 85% MIB, at 22 °C. MIB removals decreased to 60% and 46%, respectively at 16 °C and 10 °C. The application of 0.2 H_2O_2/O_3 (mg/mg) prior to BAC provided treatment which effectively removed geosmin and MIB. However, in terms of DBP precursor reduction, there was no beneficial impact of H_2O_2 addition on trihalomethane or haloacetic acid formation potentials.

1. Introduction

Geosmin and 2-methylisoborneol (MIB) have typically been reported as the most common taste and odour (T&O) causing compounds and are seasonally detected in drinking water sources as a result of anthropogenic or biogenic factors (Srinivasan and Sorial, 2011). Removal of these compounds by conventional treatment is typically low (geosmin: <20%; MIB: <15%), as such their presence in finished water is a major concern to water treatment utilities (Olsen et al., 2016; Parinet et al., 2010).

Treatment strategies designed to remove geosmin and MIB from source waters have been extensively reported (Antonopoulou et al., 2014; Srinivasan and Sorial, 2011). Adsorption by granular activated carbon (GAC) has proven to be effective for the removal of these compounds, however its efficiency is dependent on media adsorptive capacity (Gillogly et al., 1999; Ho and Newcombe, 2010). Fortunately, when the adsorptive capacity of GAC is exhausted, filters can be operated in a biological mode and continue to remove geosmin (44–87%) and MIB (32–51%), resulting in significant capital cost savings that would typically be associated with media

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regeneration or replacement (Elhadi et al., 2004; Zhu et al., 2010). In addition, BAC has been recognized for its ability to reduce biodegradable organic matter (Carlson and Amy, 2001) and disinfection byproduct (DBP) precursors (Zha et al., 2016). However, BAC performance is impacted by water temperature (Emelko et al., 2006; Moll et al., 1999), empty bed contact time (EBCT), and accumulation of attached microbial biomass (Elhadi et al., 2006). Because of these factors, BAC performance in terms of geosmin, MIB, as well as DBP precursor removals can vary significantly (de Vera et al., 2016; Elhadi et al., 2004; Guo et al., 2016).

Application of O_3 prior to biofiltration has been shown to be effective for the removal of geosmin, MIB, and DBP precursors (Klausen and Grønborg, 2010; Park et al., 2015; Sánchez-Polo et al., 2006; Yan et al., 2010). An investigation of O_3-biofiltration treatment at full-scale by Newcombe et al. (2010) reported MIB removals of 36–54% by ozonation (1.3 mg/L O_3) alone followed by 26–46% removal across BAC (17 min EBCT). Chang et al. (2002) reported reductions of 18% dissolved organic carbon (DOC), 61% UV_{254} absorbance, 17% trihalomethane formation potential (THMFP), and 19% haloacetic acid formation potential (HAAFP) when treated with 3 mg/L O_3 followed by BAC (5 min EBCT). It is important to note that O_3 efficiency is dependent on source water characteristics such as pH, DOC, alkalinity, and temperature (Westerhoff et al., 2006), and can be improved with the addition of H_2O_2 to produce hydroxyl radicals ($OH) and provide fast reaction rates for geosmin and MIB (K_{OH} ~10^9 $M^{-1}s^{-1}$) (Lee and von Gunten, 2016; Peter and Von Gunten, 2007). Previous pilot studies have reported increases of 10–15% in geosmin and MIB removals upon H_2O_2 addition (Glaze et al., 1990; Park et al., 2015; Wang et al., 2015). However, much of the existing information regarding H_2O_2/O_3 optimization has been based on the bench-scale studies (Li et al., 2012; Mizuno et al., 2011; Parinet et al., 2010; Westerhoff et al., 2006; Yuan et al., 2013; Zhou et al., 2011). Pilot-scale studies are warranted to identify an optimum range of H_2O_2:O_3 (mg/mg) ratio for this water source, which can guarantee maximum removal of T&O compounds (Acero and Gunten, 2001). Below this ratio, O_3 is present in excess, which favors O_3 decomposition over $OH formation; above this ratio, H_2O_2 is present in excess, which consumes O_3 and generates $OH.

In this study, H_2O_2/O_3 mass ratios of 0.1:1, 0.2:1, 0.35:1, and 0.5:1 were compared to O_3 alone to better understand the removal of geosmin, MIB, and organics at pilot-scale. The impact of pre-oxidation on subsequent biofiltration performance using two different media types was also assessed with respect to the same analytes, as well as organic carbon removal (measured as DOC, UV_{254}), DBP precursor removal (including precursors to THMs and HAAs), and filter media adenosine triphosphate (ATP). Liquid chromatography–organic carbon detection (LC-OCD) was performed to further characterize the impact of each treatment process and to explore potential relationships between specific organic fractions (humics or low molecular weight compounds) and DBP precursor removal.

2. Materials and methods

2.1. Source water

The Peterborough Water Treatment Plant (PWTP) is located on the Otonabee River and has a total capacity of 104 MLD. As the PWTP typically observed increased geosmin (15–22 ng/L) and MIB (10–20 ng/L) during September to November, pilot-scale studies were conducted during these months using full-scale plant settled water obtained post coagulation, flocculation, and settling. Typical pilot influent water quality during the study is shown in Table 1.

Table 1
Pilot influent (settled) water quality.

Parameter	September	October	November
pH	7.3–7.4	7.2–7.3	7.2–7.3
Temperature (°C)	20–24	16–17	9–10
Turbidity (NTU)	0.165–0.185	0.127–0.143	0.118–0.138
DOC (mg/L)	3.2–3.4	3.1–3.3	3.2–3.3
UV_{254} (cm^{-1})	0.056–0.058	0.052–0.054	0.051–0.053
Alkalinity (mg/L as $CaCO_3$)	60–62	59–60	57–60
Geosmin (ng/L)	15–23	17–21	11–16
MIB (ng/L)	18–20	11–15	BDL
THMFP (μg/L)	73.4	68.7	85.5
HAAFP (μg/L)	74.6	103.8	108.2

BDL=Below Detection Limit (Geosmin = 5.7 ng/L; MIB = 7.1 ng/L).

2.2. Pilot plant design and operation

Six pilot-scale filters were operated in parallel at an EBCT of 10 min to mimic full-scale conditions (Table 2). Three filters contained 50 cm of sand (effective size (d_{10}) = 0.5, uniformity coefficient (UC) = 1.8) and 50 cm of biological exhausted GAC (Filtrasorb® 300 Calgon Carbon; Pittsburgh, PA; d_{10} = 0.8; UC = 2.0) that had been in operation for 12 years and was assumed to be exhausted in terms of adsorptive capacity. The remaining three filters contained 50 cm of sand (d_{10} = 0.5, UC = 1.8) and 50 cm of anthracite (d_{10} = 0.85, UC = 1.8), which was obtained from the full-scale PWTP (Table 2). All filters were operated in a constant head and declining flow mode. In order to maintain conditions conducive to biological filtration, five of the pilot filters were backwashed with their individual (unchlorinated) effluent, while the conventional pilot filter was backwashed with chlorinated water (chlorine residual = 1.5 mg/L) from the full-scale plant (FSP). All filters were backwashed three times a week, at 50% filter bed expansion for 8 min followed by 4 min of slow backwash (20% fluidization) to maintain proper media layer separation (Azzeh et al., 2015).

The O_3 pilot consisted of two parallel trains (4 contactors of 65 L each, with 2 contactors connected in series), supplied with full-scale settled water, and operated at a flow rate of 11 L/min to achieve an O_3 contact time of 12 min. Based on O_3 demand tests conducted using settled water, 2 mg/L O_3 was applied. The transferred dosage of O_3 in both the trains averaged 2.04 ± 0.09 mg/L at an average transfer efficiency of 97.8%. The selected dose corresponded to a O_3:DOC ratio of 0.625, and within an operational range of O_3 dosages (O_3:DOC = 0.5 to 1) as described previously for optimal organics removal (Schulz, 2014; Zimmermann et al., 2011). An influent concentration of 100 ng/L was targeted for geosmin and MIB spiking trials (Ferguson et al., 1990; Liang et al., 2014). Spiking was conducted for 24-h periods using a Masterflex L/S pump; geosmin and MIB samples were collected 24 h after each spiking event to simulate transient T&O events. H_2O_2 was added immediately prior to O_3 to obtain mass H_2O_2:O_3 ratios of 0.1:1, 0.2:1, 0.35:1, and 0.5:1 mg/mg. Residual oxidants were quenched at the exit of the ozone pilot using calcium thiosulphate (CTS) (30% w/v) at a molar ratio of CTS:O_3 (mol/mol) = 4:1. Stoichiometric ratio for CTS:O_3 (mg/mg) was 10.5:1, i.e. for every gram of O_3, 10.5 g of CTS (30% w/v) was required.

2.3. Experimental design and sampling procedure

This study considered four experimental conditions (Table 2). For condition 1, geosmin, MIB, and DBP precursor removals were evaluated across the conventional anthracite filters (CAF) and biological anthracite filter (BAA) and biologically active carbon filters (BAC) without the use of pre-oxidation. For condition 2, the impact of 0.1 H_2O_2/O_3 and O_3 were compared as stand-alone treatments